Containerless Experimentation in Fluid Dynamics and Crystal Growth on Earth and in Microgravity

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The experimental techniques used for the investigation of the shape dynamics and flows within single isolated fluid particles have been refined to the point where detailed differences between phenomena observed in 1 G and in microgravity can be accurately measured. Recent experimental results have been obtained concerning the nonlinear dynamics of free drops and bubbles in 1 G and in microgravity. Specific indirect effects of gravity on the nonlinear dynamics of shape oscillations of bubbles have been identified and the detailed evaluation of a single particle ultrasonic positioning system has been carried out in the NASA Glovebox facility in microgravity. These results imply that the enhanced symmetry offered by the microgravity conditions would yield qualitatively different results for bubbles undergoing large-amplitude oscillations within a restraining field. Recent flight investigation results also suggest that very simple ultrasonic positioning devices could be used in low-gravity to carry out accurate measurements of phenomena associated with single isolated droplets. Finally, experiments in 1-G using an ultrasonic-electrostatic hybrid system are being carried out to gather information regarding buoyancy-coupled thermocapillary flows within a levitated laser-heated drop, and about the protein crystal growth process in levitated solution drops rotating along a horizontal axis.

1. Introduction

Experimentation in microgravity not only benefits from the drastic reduction of natural buoyancy found on Earth, but also from the opportunity to carry out truly containerless investigations. It is now clear that field effects associated with single fluid particle levitation in 1-G significantly influence the dynamics and the internal flow regime of the samples under investigation. This interfering action leads to both qualitative and quantitative deviations from phenomena that would be observed in the absence of levitation field restraints. An obvious and vivid example of the imposed field effects is the equilibrium shape deformation of levitated drops and trapped bubbles in 1-G. It has been both theoretically and experimentally established that a distortion of the equilibrium shape of a drop or bubble from the spherical geometry leads to a shift in the resonant frequencies of natural shape oscillations[1,2]. Another well recognized artifact is the induced internal flows found within electromagnetically levitated conducting melts. Such circulation also impacts the natural shape oscillations of these drops, as suggested by a recent theoretical modeling effort[3]. Thus, it is clear that any application based on the measurement of the oscillatory responses of drops under these field constraints should carefully include them in the analysis.

Although numerical modeling and theoretical analysis of the fluid dynamical aspects of field effects both provide valuable guidance, the experimental verification of their predictions is required. To this end, investigations using Earth-based simulations of low-gravity conditions and

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actual low-cost microgravity experiments can be rationally justified. In this paper, we describe some of our results obtained through the development of containerless methods for Earth-bound as well as microgravity-based laboratories. The ultimate motivation of the described research is the applications of these techniques to the understanding of phenomena relevant to Earth-based technologies as well as to the technical problems of manned or robotic Space exploration.

In the first part of the paper we discuss some of the aspects of single drop and bubble dynamics that are indirectly influenced by gravity using experimental results obtained at 1-G and in microgravity. In the second part, we address new containerless methods used in the investigation of thermocapillary flows and protein crystallization in levitated and rotating drops. The emphasis of this review paper is to succinctly describe some of the currently available containerless techniques and their application to specific problems in fluid dynamics and crystal growth.

2. Drop and Bubble Dynamics at 1-G and in Microgravity

Ultrasonic levitation methods are particularly well suited for both Earth-based as well as Microgravity experimentation because the magnitude of the acoustic radiation stresses can be continuously adjusted to conform to the level of gravitational influence. In other words, the interfering side effects can be reduced to virtually zero in a quiescent low-gravity environment. For example, in the case of a single-axis ultrasonic levitator[4-7], the positioning of a 5 mm diameter water droplet in air at 1-G invariably causes a static equilibrium shape deformation into an approximately oblate spheroid, drives an uncontrolled drop spinning motion, and induces internal flow. All these side effects can be reduced to a very low level by decreasing the acoustic power level. **Figure 1** shows a series of photographs of a 5 mm diameter levitated water droplet at different acoustic intensity and at 1.8 G, 1.0 G, and 0.05 G effective gravity levels obtained in the NASA KC-135 airplane flying parabolic trajectories. At the lowest G-level, a virtually spherical droplet can be obtained while still retaining active acoustic positioning. In this particular instance, the ultrasonic frequency was 37 kHz, and the droplet diameter was a substantial portion of the acoustic half-wavelength.

A similar ultrasonic device designed for liquid-filled resonant cavities allows the trapping of relatively large gas and vapor bubbles (0.5 to 10 mm diameter) in a liquid host. As for the case of droplet levitation, the acoustic radiation pressure used to trap a bubble also distort its equilibrium shape. This distortion becomes more pronounced as the size of the bubble relative to the acoustic wavelength increases.

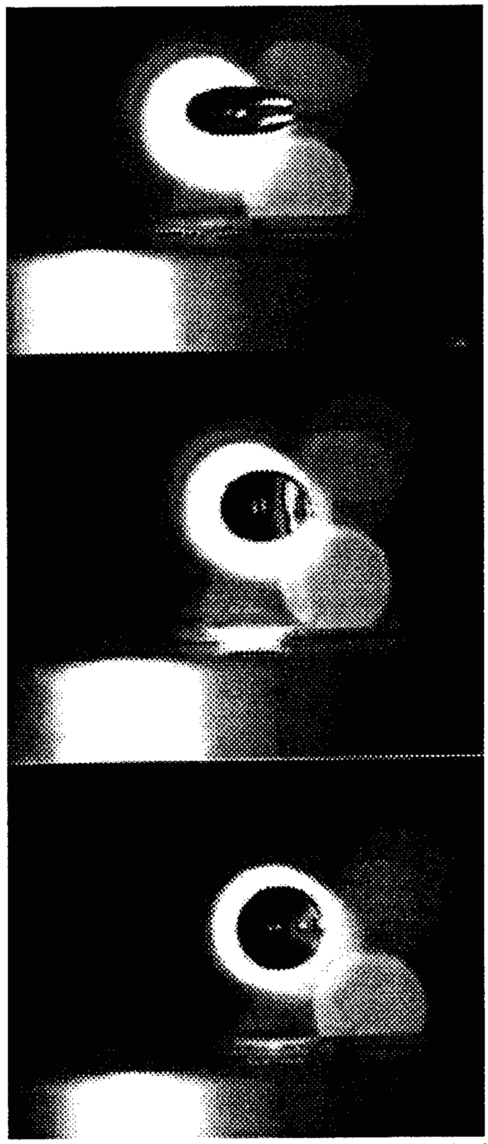

Figure 1 The shape of an acoustically levitated 5 mm diameter water droplet at 1.8, 1.0, and 0.05 G.

Gravity causes the bubble to be trapped in a position slightly above an acoustic pressure nodal plane in the standing wave. This cause an asymmetry in both the static and dynamic shape. In addition, the coupling of the acoustic oscillations to the bubble surface results in the onset of capillary waves.

As shown in **Figure 2**, this distortion in the static shape influences the dynamics of bubble shape oscillations. From these results, it is clear that bubbles having a radius larger than about 3 mm will yield measured resonance frequencies higher than the theoretical predictions based on linear theory. Measurement in low-gravity with even larger, but undistorted bubbles gives values which agree with the theoretical results[8]. This shift in frequency arises from two causes: the first is due to the shape distortion itself, and the second comes from the acoustic radiation pressure which combines with surface tension and alters the surface restoring force. Shape oscillations resonant frequencies are frequently used to infer the surface tension at the liquid-gas interface, and ignoring these finite size effects would therefore lead to erroneous values.

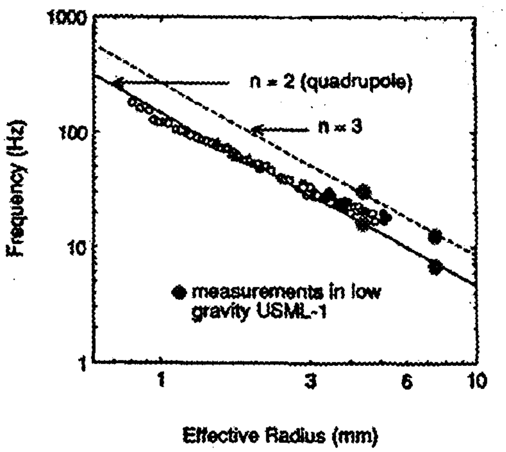

Figure 2. The measured resonance frequencies for the first two resonant modes of bubble shape oscillations (Marston et al., 1994).

The shape of bubbles undergoing large-amplitude shape oscillations is also influenced by asymmetrical position of the bubble relative to a pressure nodal plane: At 1-G (or high Gs), the prolate shape characteristic of the fundamental quadrupolar mode has a rounded north pole and a pointed south pole as shown in **figure 3**. This becomes more accentuated as the oscillation amplitude is increased. **Figure 4** shows this asymmetrical motion even more clearly. This distortion will significantly affect the results of an analysis of the dynamic shape using modal decomposition: a larger proportion of higher mode shapes would be detected, and this result would not only reflect any nonlinear coupling phenomenon, but also an asymmetrical ultrasonic force field. Experimental evidence also shows, however, that this asymmetry does not exist in a low-gravity environment where the shape oscillations are symmetrical even at large amplitude.

Figure 3. Shapes of a bubble undergoing shape oscillations in the fundamental quadrupolar mode under strobed illumination. The left shape is for 1-G conditions while the right shape is obtained in low-gravity.

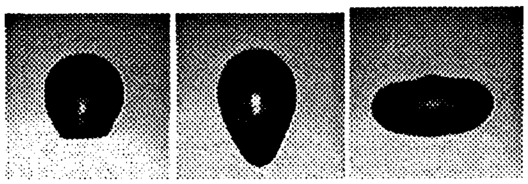

Figure 4 High-speed video frames (4000 fps) of an oscillating bubble displaying the drastic asymmetrical motion of the upper and lower hemispheres. This asymmetry has not been observed under low-gravity conditions.

The nonlinear shape oscillatory response of both single drops and bubbles to an asymmetric acoustic radiation pressure excitation shows the characteristics of a soft weakly nonlinear system. The natural (free-decay) resonance frequency of the fundamental quadrupolar mode has been theoretically predicted[9-11)] and experimentally shown[12,2)] to decrease with increasing oscillation amplitude. Because the influence of the levitation

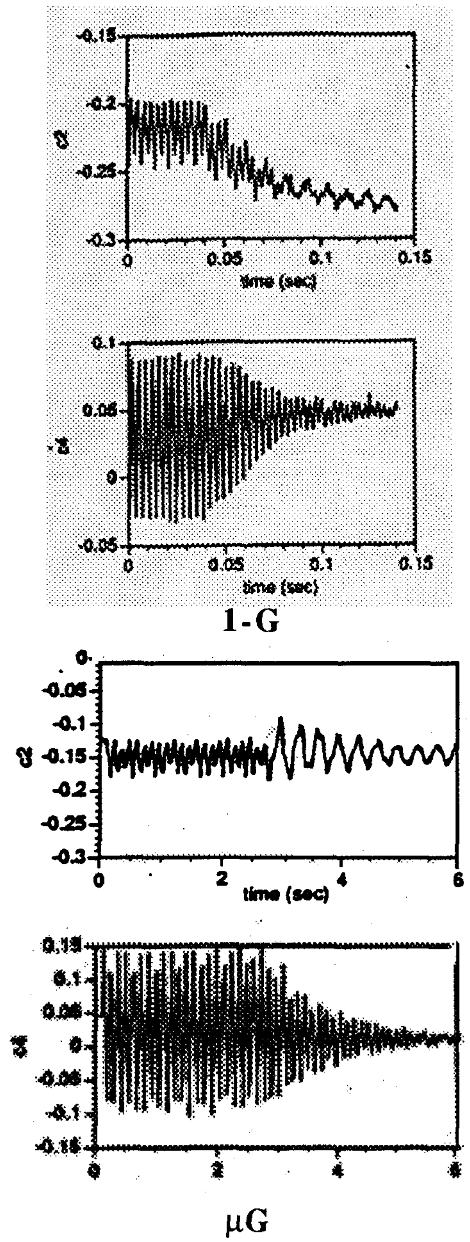

Figure 5. Free -decay of levitated drops in 1-G and in microgravity both initially driven into the resonant L=4 mode oscillations. The major difference is the immediate onset of the L=2 mode oscillations upon termination of the electric field drive for the 1-G case and of acoustic oscillation drive for the μG case. The C_2 and C_4 coefficients are the Legendre coefficients used in the expansion of the drop shape into Legendre polynomials.

field is a significant factor in the detailed dynamics of the fluid particles, low-gravity measurements performed in the absence of a restoring external force field are required. Some of this type of information has been obtained during low-gravity investigations both on the KC-135 airplane and during a Shuttle flight investigation using the a Spacelab facility (Drop Physics Module)[13] and a small Glovebox experiment[14]. The results indicate that, within the currently obtainable experimental accuracy, the effects of the acoustic restoring force does not appear to drastically alter the conclusion that the natural fundamental mode resonant frequency decreases with increasing oscillation amplitude.

A difference between Earth-based and micro-gravity results has been observed, however, when a drop is initially oscillating in an axisymmetric higher mode. In one instance, the free decay regime of a drop initially driven into the L=4 mode has been examined for a drop levitated on Earth and for one investigated in microgravity. **Figure 5** displays the results of the modal analysis with only the L=2 and L=4 oscillations shown. The analysis of axisymmetric drop shape oscillations usually involves the expansion of the shape in terms of Legendre polynomials:

$$r(\theta) = R_0 + \sum_{n=2}^{m} C_n P_n(\cos\theta)$$

where θ is the polar angle, P_n is the Legendre polynomial of order n, C_n is the coefficient of P_n, and m is the maximum order used in the fitting procedure. The graphs of **Figure 5** are only for the C_2 and C_4 coefficients.

The drop observed in microgravity exhibits an immediate excitation of the L=2 mode upon termination of the active oscillation drive, while the Earth-based drop still shows substantial coupling between the L=2 and L=4 modes in the free decay region. One might conclude that experimentally observed nonlinear mode coupling phenomena found at 1-G are more directly related to the levitation field effects. This is clearly reflected by the fact that eliminating the active oscillation drive reduces the static deformation for both the L=2 and L=4 shapes (the equilibrium static deformation shifts to different values during the free-decay phase).

Having clearly established that the constraining effects of the levitation and manipulation fields influence the response of the fluid particles, one should, therefore, anticipate that microgravity conditions would allow the calibration of these interfering characteristics. An understanding of the field-induced phenomena will greatly improve the applicability and value of containerless material characterization and processing methods in 1-G.

In the remainder of this paper, we will address some recently developed experimental capabilities for both microgravity and 1-G studies in fluid dynamics and crystal growth. We will describe a versatile and low-cost Glovebox-based experimental apparatus and two ground-based investigations in thermocapillary flows and protein crystal growth in simulated low-gravity.

3. Microgravity Glovebox Apparatus and Experiments

We have developed a compact, low-cost, and versatile apparatus to carry out controlled and quantitative experiments on single free drops and bubbles within the NASA Glovebox facility. It has been used to test the feasibility of quiescently positioning liquid and gas samples using acoustic radiation pressure; the crucial information obtained was the effect of the ultrasonic power level on the translational and rotation stability of a liquid drop in air and the positioning effectiveness of a gas bubble in a liquid host in actual microgravity conditions during a Space Shuttle orbital flight. Quantitative measurements of the dependence of oscillation dynamics on the magnitude of the ultrasonic field are also feasible by using a simple light extinction measurement method provided by a collimated diode laser and silicon detector apparatus connected via a PCMCIA interface to a Laptop computer.

The apparatus shown in **Figure 6** was used during the STS-94 flight in 1997 to carry out an investigation titled The Internal Flows in Free Drops (IFFD). The fundamental goal was to investigate the accuracy with which a single-axis ultrasonic levitator could position a free drop in air. The primary parameter of interest is the residual rotational velocity of a freely deployed droplet in the Shuttle environment. The behavior of the drop was monitored using laser light scattered from suspended tracer particles. The motivation was provided by the measurement of thermocapillary flows within a free, spot-heated drop, the configuration of which had already been theoretically analyzed[15] . The implementation of these measurements in 1-G using an Ultrasonic-Electrostatic hybrid levitator is discussed later in this paper.

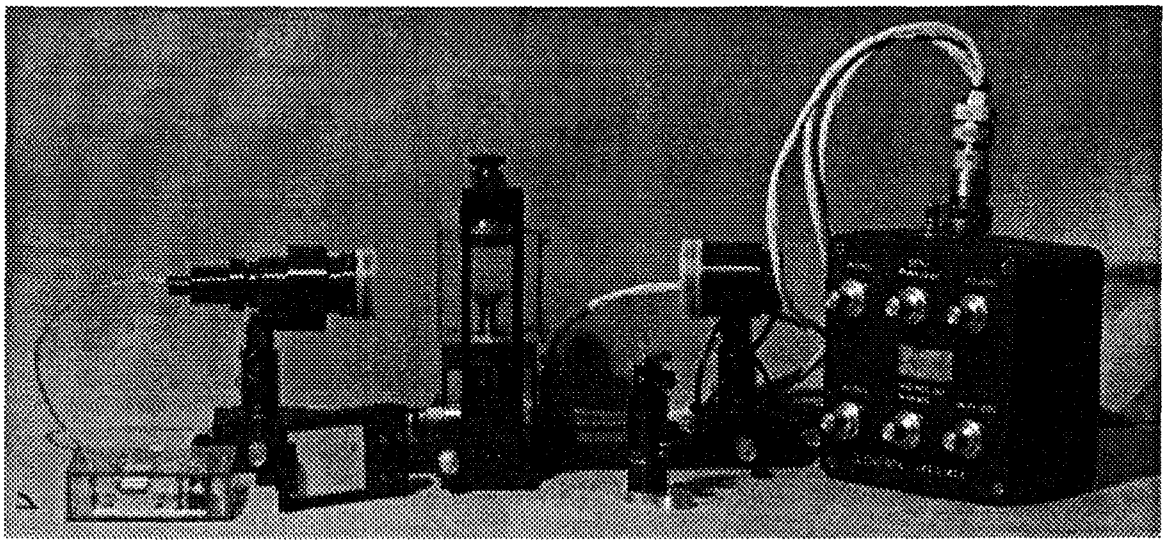

Figure 6. IFFD apparatus used on STS-94 for the study of the stability of free drops and of the internal flows under isothermal conditions. The diode laser (extreme left) powered by AA batteries is used to illuminate the drop to visualize the motion suspended tracer particles within drops positioned by the single-axis levitator (center). The dynamics of the drops could also be measured through optical extinction by the photodetector (right) connected via a PCMCIA interface to a laptop computer. The levitator is manually controlled by the Electronic Control Unit (extreme right). The apparatus was operated in the NASA Middeck Glovebox facility.

The single-axis ultrasonic device operates at 21 kHz, and can accommodate droplets having a diameter up to 1 cm, although only 5 mm-diameter droplets were used in the recent microgravity investigation. The major challenges of this totally crew-operated experiment were the drop deployment and stabilization phases. The injection and launching of the free drops were manually performed by the operator: the acoustic force capturing the deployed drop within a force field is used to detach the droplet from the deployment needle and to position it within the stable potential well. Because the main purpose of the investigation was to measure the lowest possible force field allowable, periods of time free of residual acceleration were required. Crew motion and mechanical device activation within the Shuttle could easily disturb the droplet from its equilibrium position.

An acoustically levitated droplet is driven into rotation by the air convection currents induced by the high-intensity sound field. These steady convective flows are second order effects, and are directly generated by the alternating air motion generated by the sound field as it interacts with the sample and any other boundaries. The levitated drop is driven into rotation due to aerodynamic drag. The axis of rotation is generally uncontrolled, but can be directed along a fixed direction using mechanical adjustments in the levitator. In the particular device used in this Glovebox apparatus, the rotation axis is adjusted to be horizontal and perpendicular to the levitator symmetry axis.

Figure 7 displays the recorded residual rotational stability of two different droplets during the Shuttle flight as a function of the ultrasonic power setting. It is evident that residual uncontrolled rotation along a fixed axis as low as 0.2 rps was obtainable for the approximately 5 mm-diameter drops of glycerin-water mixtures. A lower rotational velocity could be potentially obtained for longer waiting time due to low drag exerted by the ambient air on the slowly rotating droplet.

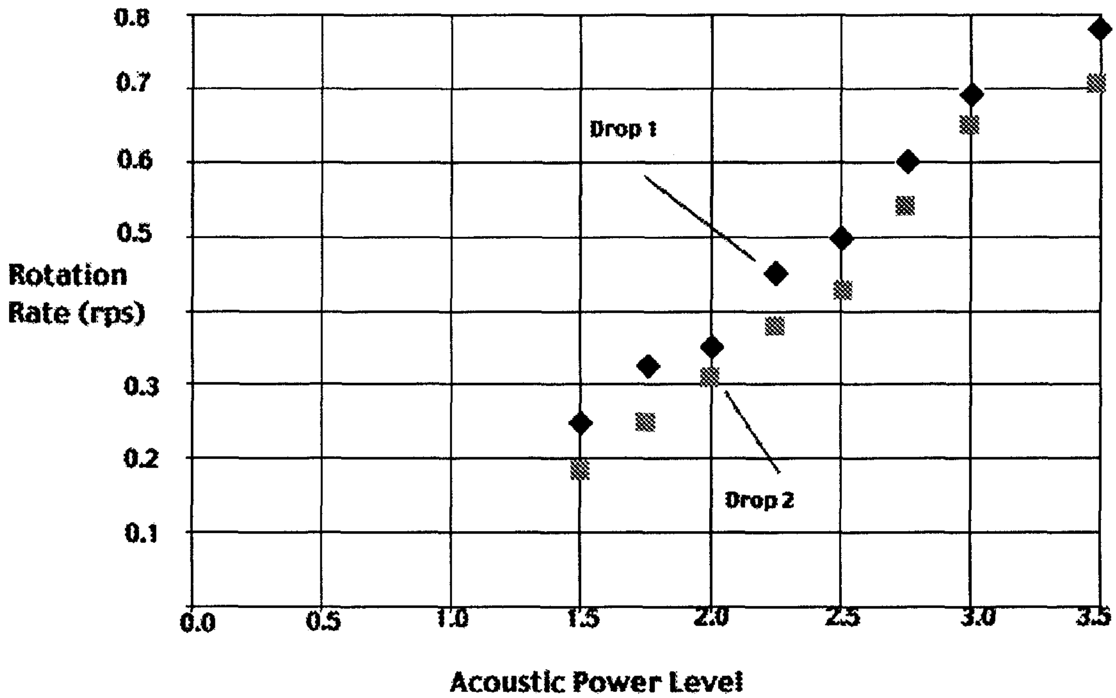

Figure 7. The measured evolution of the rotational velocity of a free droplet positioned in the IFFD levitator during the STS-94 flight. The residual rotation was measured as a function of the acoustic power level for two different drops of water-glycerin mixture. The rate was measured by tracking suspended tracer particles illuminated by a collimated diode laser. Levitation of smaller volume droplet in 1-G would require a drive level on the order of 7.0. Lower residual rotation could be obtained for longer observation times because of the low drag exerted by the ambient air.

4. Protein Crystal Growth in Levitated Rotating Drops and Thermo-capillary-Buoyancy driven Flows in Laser-heated Free Drops in 1-G.

Recent advances in containerless experimentation methods are creating new opportunities for innovative scientific and technological investigations. Two specific areas of particular interest to the Microgravity Science program are: 1) The influence of gravity and fluid flow on the protein crystallization process and 2) Thermocapillary convection in droplets. We briefly describe below our recent progress in the application of containerless methods to those problems.

By combining the capabilities of ultrasonic and electrostatic levitators, it has been possible to implement a unique approach to protein crystallization that allows the control of heterogeneous nucleation, the simulation of low-gravity conditions, the investigation of the effects of fluid convection in a controlled manner, and the accurate monitoring and control of the growth conditions. By electrostatically levitating charged droplets of protein solution and using the ultrasonically-induced torque over periods of several days, we have been able to growth large crystals (300 μm to 1 mm) of Lysozyme and Thaumatin in 1-G[16,17]. In the case of Thaumatin, we have observed a great reduction in the nucleation rate when compared to standard hanging and sitting drop methods in the same growth conditions. In spite of a significant density difference, the long-term suspension of large crystals (greater than 200 μm) within the bulk of the solution droplets has been made possible by using very dilute solutions (less than 0.01%) of agarose. Under steady-state and isothermal growth conditions, we have carried out the observation of the fluid flow within the rotating droplets in 1-G, and recorded a slow drifting motion. By measuring the motion of growing crystals within the rotating levitated solution droplets we have found that the crystals were drifting at a slower rate than that recorded in Shuttle-based experiments using contained solution growth.

Figure 8 shows crystals of Lysozyme and Thaumatin suspended within levitated and rotating droplets in 1-G. A significant advantage of this approach is derived from the ability to introduce internal flows within the solution in a controlled manner by driving drop shape oscillations. A quantitative study of the effects of convection can therefore be obtained.

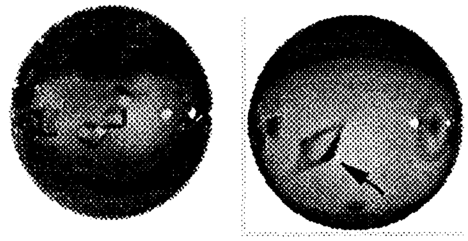

Figure 8. Levitated solution droplets containing Lysozyme (left) and Thaumatin crystals grown in 1-G within levitated and rotating droplets.

In coordination with the IFFD Glovebox flight investigation, we have also initiated the ground-based experimental study of the internal flows within electrostatically levitated droplets in isothermal conditions as well as under focused laser heating. Using stereoscopic imaging based on two closely spaced video cameras, we have been able to observe the onset on thermocapillary-buoyant flows within initially quiescent droplets in 1-G. The laser beam is horizontally-directed to impact a small area (70 μm diameter) at a point on the drop equator. Preliminary experiments have used low-power pulses from a focused CO_2 laser with an modest temperature increase (less than 30 °C). Suspended tracer particles allow the visualization of the internal flows. **Figure 9** displays video frames used for the simulated 3-D imaging of the flows within a levitated and spot-heated droplet.

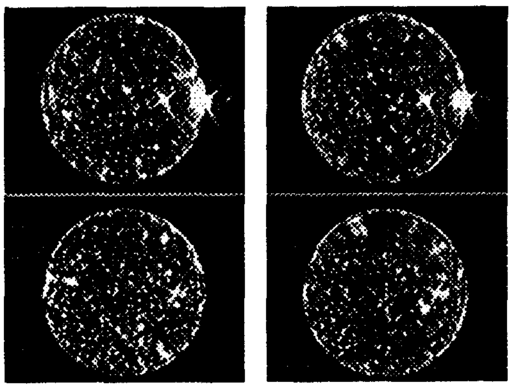

Figure 9. Video frames of levitated drops under spot-heating form a focused laser. The images from two slightly displaced cameras allow a simulated 3-D flow visualization. Ray tracing correction must be used to compensate for the drop curvature, and

digital analysis allows the reconstruction of the flow field.

Summary.

Containerless experimentation methods have been refined and have been adapted to experimental investigations in a variety of scientific disciplines of relevance to the Microgravity Research program. The judicious combination of containerless methods with the low-gravity environment should allow the calibration of interfering field effects and the development of rigorous and accurate thermophysical properties measurement methods to be used in 1-G. In addition, the opportunity offered by low-gravity will allow the controlled and precise experimental studies of a wide variety of physical phenomena dominated by capillary forces and of the effects of gravity on crystal growth processes of practical significance.

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